<International Research Center for Elements Science>Nanophotonics

International Research Center for Elements Science – Nanophotonics –

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Scope of Research

Our research interest is to understand optical and quantum properties of nanometer-structured materials and to establish opto-nanoscience for creation of innovative functional materials. Space- and time-resolved laser spectroscopy is used to study optical properties of semiconductor quantum nanostructures and strongly correlated electron systems in low-dimensional materials. The main subjects are as follows: 1) investigation of optical properties of single nanostructures through the development of a high-resolution optical microscope, 2) ultrafast optical spectroscopy of excited states of semiconductor nanostructures, and 3) photophysics of solar cell materials.

KEYWORDS

Femtosecond Laser Spectroscopy
Single Photon Spectroscopy
Semiconductor Nanoparticles
Solar Cells
Perovskites

Selected Publications

Handa, T.; Tahara, H.; Aharen, T.; Kanemitsu, Y., Large Negative Thermo-optic Coefficients of a Lead Halide Perovskite, *Sci. Adv.*, **5,** [eaax0786-1]-[eaax0786-8] (2019).

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Tahara, H.; Sakamoto, M.; Teranishi, T.; Kanemitsu, Y., Quantum Coherence of Multiple Excitons Governs Absorption Cross-Sections of PbS/CdS Core/Shell Nanocrystals, *Nat. Commun.*, **9,** [3179-1]-[3179-8] (2018).

Yamada, T.; Aharen, T.; Kanemitsu, Y., Near-Band-Edge Optical Responses of $CH_3NH_3PbCl_3$ Single Crystals: Photon Recycling of Excitonic Luminescence, *Phys. Rev. Lett.*, **120,** [057404-1]-[057404-6] (2018).

Tahara, H.; Sakamoto, M.; Teranishi, T.; Kanemitsu, Y., Harmonic Quantum Coherence of Multiple Excitons in PbS/CdS Core-Shell Nanocrystals, *Phys. Rev. Lett.*, **119,** [247401-1]-[247401-6] (2017).

Negative Thermo-optic Coefficient in Metal Halide Perovskites

Metal halide perovskites have been the subject of considerable research efforts due to their potential applications in optoelectronics. Understanding the refractive index and its change upon temperature rise is very important for the optimum design of semiconductor photonic devices. We found that the halide perovskite $CH_3NH_3PbCl_3$ shows a decrease in the refractive index with temperature, *i.e.*, negative thermo-optic coefficient. This negative thermo-optic coefficient is opposite to the positive coefficients in conventional inorganic semiconductors. Therefore, the halide perovskite works as a compensator of temperature-induced optical phase shift that inevitably occurs in inorganic semiconductors. To prove this compensation, we measured the optical phase shifts for conventional inorganic semiconductor ZnSe with and without $CH_3NH_3PbCl_3$ compensator (Figure 1(a)). As shown in Figure 1(b), we found that $CH_3NH_3PbCl_3$ actually compensates the optical phase shift of ZnSe. This discovery of the negative thermo-optic coefficient is of great importance for practical applications and adds new functionalities to the halide perovskites.

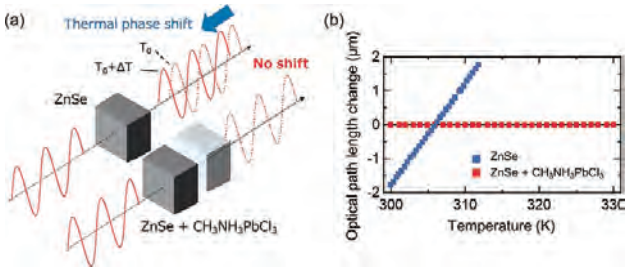

Figure 1. (a) Schematic of compensation measurement of optical phase shift. (b) Experimental results of optical phase shifts.

High Harmonic Generation from Metal Halide Perovskites

Light with a much higher photon energy than an input photon energy is generated by illuminating a solid with intense pulse laser. This phenomenon is called high-order harmonic generation (HHG). HHG originates from nonlinear currents that are caused by strong driving of electrons and holes by a high-intensity electric field, but the detailed mechanism is still unclear. In addition, exploration of materials with high generation efficiency is necessary from the viewpoint of photonic device application. We observed high efficiency HHGs up to the 13th order from organic-inorganic hybrid perovskite [$MAPbX_3$ (MA = CH_3NH_3, X = Cl, Br, I)]. Here, the mid-infrared laser pulse centered at energy of 0.35 eV with a maximum peak electric field of 10 MV/cm was used for photoexcitation. This finding is quite important for application to HHG light sources, since halide perovskite thin films are easily fabricated over a large area.

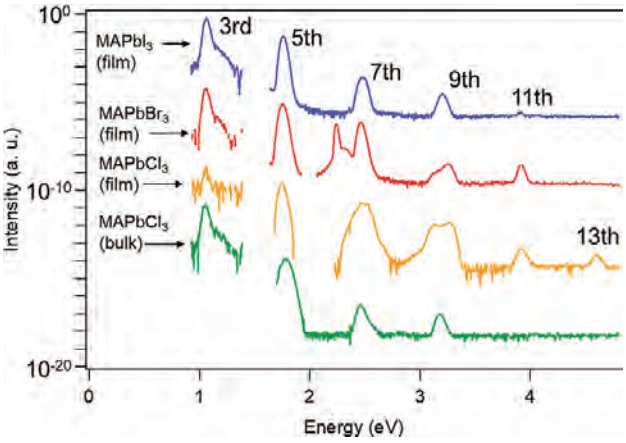

Figure 2. High harmonic generation spectra from methylammonium lead halide perovskite thin films.

Ionization and Neutralization Processes in Metal Halide Perovskite Nanocrystals Studied by Double Pump Spectroscopy

Lead halide perovskite $CsPbX_3$ (X=Cl, Br, I) nanocrystals have excellent properties such as very high luminescence efficiencies and efficient optical gain, then, are expected for various light-emitting device applications. Trions (charged excitons) in nanocrystals have a disadvantage of reducing luminescence efficiencies, but also have an advantage of lowering the optical gain threshold. Therefore, a detailed understanding of the trion generation and relaxation dynamics is required for device applications. We studied the ionization and neutralization processes of $CsPbBr_3$ nanocrystals by using double-pump transient absorption spectroscopy. It is clarified that photo-ionization processes are attributed to the radiative and non-radiative Auger recombination of biexcitons and trions, and neutralization processes are governed by the fast (~990 ps) and slow (~12 μs) process (Figure 3).

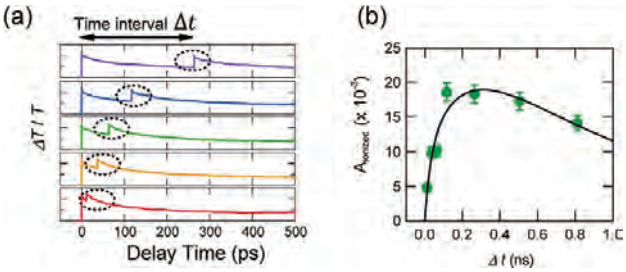

Figure 3. (a) Double pump transient absorption signals for various time intervals. (b) Time interval dependence of signal amplitude of ionized nanocrystals.